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Title: **STUDY OF THE IDGS TECHNIQUE FOR MIXED PLUTONIUM-URANIUM (MOX) SAMPLES**

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(ABSTRACT)

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Study of the IDGS Technique for Mixed Plutonium-Uranium (MOX) Samples

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ABSTRACT

The isotope dilution gamma-ray spectrometry (IDGS) technique has been demonstrated for simultaneously measuring concentrations and isotopic compositions of plutonium in spent-fuel input dissolver solutions. For timely analyzing nuclear materials on the purpose of material accountancy and quality control/assurance, we have performed a feasibility study to implement the IDGS for measuring mixed plutonium-uranium oxide (MOX) samples at the Plutonium Fuel Center (PFC) of Japan Nuclear Cycle Development Institute (JNC). Proof-of-principle experiments and analysis have been conducted for developing simultaneous plutonium and uranium measurements in MOX samples with wide variation of Pu/U ratios including powder, pellets and process scraps from the MOX fuel fabrication plant at PFC.

1. INTRODUCTION

In the IDGS method, the concentration of plutonium in the unknown solution is determined by calculating the differences among the isotopic $^{240}Pu/^{239}Pu$ ratios of the spike, the spiked solution, and the unknown solution.

$$C(Pu) = \frac{M_s}{V_u} \bullet \frac{W_s^9}{W_u^9} \bullet \frac{R_m - R_s}{R_u - R_m}, \qquad (1)$$

where

M_s = mass of plutonium in the spike
V_u = volume of dissolver solution taken
W_s^9 = weight fraction of ^{239}Pu in the spike
W_u^9 = weight fraction of ^{239}Pu in the unknown solution
R_m = the $^{240}Pu/^{239}Pu$ ratio in the mixed or spiked solution
R_s = the $^{240}Pu/^{239}Pu$ ratio in the spike
R_u = the $^{240}Pu/^{239}Pu$ ratio in the unknown solution

In this equation, the values of M_s, V_u, W_s^9, and R_s are known. Only the values of R_u and W_u^9 in the unknown solution and R_m in the spiked solution need to be measured by gamma-ray spectrometry [1,2].

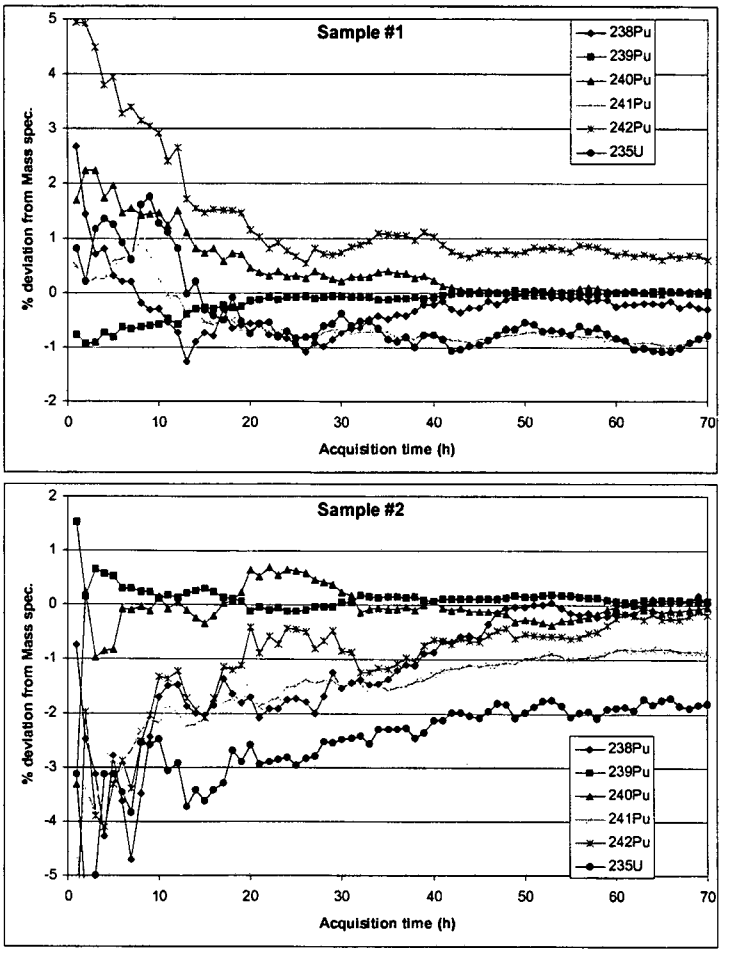

Figure 1. Deviation of the FRAM analytical results from Mass spectrometry results.

From figure 1, we can see that the isotopic results behave as expected: they scatter wildly from the expected values when the acquisition times are small (weak statistics) and they all converge to the expected values as more data are obtained, including ^{242}Pu. Note that ^{242}Pu is determined from correlation and is not expected to be very accurate. We did not modify the ^{242}Pu correlation for these data and it is just coincidence that the ^{242}Pu results agree excellently with the MS results. For typical measurements, we may expect the systematic error for ^{242}Pu correlation to be about 5-10%.

Table 2 shows the ratio of the gamma rays spectrometry results to that of the MS results of the 70-h spectra, including the FRAM's reported uncertainties.

We see that the results of ^{238}Pu, ^{239}Pu, and ^{240}Pu agree well with the MS results, well within the uncertainties. The errors for ^{242}Pu are from statistics only and do not include the systematical errors from correlations. From other FRAM analyses, we know that the FRAM reported uncertainty for ^{241}Pu is about a factor of two too small. The true errors for ^{241}Pu for the results in Table 2 should be about 0.5% instead of 0.20% or 0.25% as shown. With 0.5% uncertainty then the ^{241}Pu results would agree with that of the MS within 2 sigmas.

Table 2. Ratios of FRAM's isotopic results to that of the mass spectrometry for the 70-h spectra. The errors are the 1 sigma uncertainties reported by FRAM.

Samples	Pu238		Pu239		Pu240		Pu241		Pu242		U235	
	FRAM / MS	% error	FRAM / MS	% error	FRAM / MS	% error	FRAM / MS	% error	FRAM / MS	% error	FRAM / MS	% error
#1	0.9971	0.46	1.0003	0.10	0.9998	0.27	0.9914	0.20	1.0060	0.47	0.9922	0.48
#2	1.0002	0.58	1.0006	0.12	0.9996	0.34	0.9912	0.25	0.9982	0.59	0.9818	0.60

As for the ^{235}U results, where the results are several sigmas away from the MS results, it could very well be that the 2 sets of branching ratios for plutonium and uranium used in FRAM are

materials from different sources. That is, each plutonium material from a type of reactor (BWR, PWR, CANDU, etc.) or reactor burning different fresh fuel (uranium or MOX) will have the ^{242}Pu correlations optimized for that material instead of the current one-size-fit-all ^{242}Pu correlation formula.

CONCLUSION

We have shown that FRAM can be used with the IDGS technique to simultaneously determine plutonium and uranium isotopic compositions and concentrations in MOX samples at PFC, JNC. The uncertainties of the results are somewhat large due to weak statistics. If better statistics are obtained by either using more plutonium in the measurements, acquire the data for longer time, or using higher efficiency detector then the results can be better. The accuracy of the results can also be improved by a factor of 2-3 by using the generalized IDGS technique [7] instead of this traditional IDGS.

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